Magnetotransport properties of CeRu₂Al₁₀: Similarities to URu₂Si₂

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We report on magnetotransport properties of the Kondo semiconducting compound CeRu₂Al₁₀, focusing on its exotic phase below $T_0 = 27$ K. An excess thermal conductivity κ emerges below T_0 and is gradually suppressed by magnetic field, strikingly resembling those observed in the hidden-order phase of URu₂Si₂. Our analysis indicates that low-energy magnetic excitation is the most likely origin, as was also proposed for URu₂Si₂ recently based on inelastic x-ray scattering measurements of phonon dynamics, despite the largely reduced magnetic moments. Likewise, other transport properties such as resistivity, thermopower, and Nernst effect exhibit distinct features characterizing the very different charge dynamics above and below T_0 , sharing similarities to URu₂Si₂, too. Given the exotic nature of the ordered phase in both compounds, whether a unified interpretation to all these observations exists appears to be extremely interesting.

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CeRu₂Al₁₀ is one of the heavy-fermion (HF) materials showing timely interest due to a novel phase transition at an abnormally high temperature $T_0 = 27$ K emerging in a Kondo semiconducting phase [1-3]. An antiferromagnetic scenario, as originally proposed by one of the authors [1] based on magnetic and thermodynamic measurements, has been frequently argued to be incompatible with the large nearest neighbor distance between Ce ions, \sim 5.2 Å, and the de Gennes scaling of ordering temperatures of RRu_2Al_{10} (R = rareearth). μ SR and neutron powder diffraction experiments have revealed a long-ranged, collinear antiferromagnetic ordering of the Ce sublattice below T_0 with, however, a strongly reduced magnetic moment of 0.34–0.42 $\mu_{\rm B}$ and an unusual alignment along the *c* axis that is not the magnetic easy direction [3-5], i.e., the direction showing the largest susceptibility and magnetization. The microscopic origin of the magnetic ordering, which apparently goes beyond the conventional Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, remains elusive. On the other hand, transport properties of CeRu₂Al₁₀ are intriguing as well. For instance, regardless of the formation of a partial gap over the majority of the Fermi surface with the onset of the new phase, the electrical resistivity $\rho(T)$ ceases to be semiconductinglike and becomes metal-like upon cooling below T_0 . Furthermore, both thermopower S(T) and thermal conductivity $\kappa(T)$ are accompanied by an extra peak at temperatures slightly below T_0 or deep inside the ordered phase [1,6].

In this paper, we investigate the magnetotransport properties of CeRu₂Al₁₀. Special attention is put on the excess thermal conductance emerging in the ordered phase below T_0 and its strong suppression upon applying magnetic field. Note that strikingly similar behaviors have been observed for another heavy-fermion system, URu₂Si₂, in the hidden-order phase below $T_h = 17.5$ K [7,8]. There, enhanced phononic thermal conductance arising from a freezing out of relevant scattering centers below T_h has been argued to be the origin. This argument, however, is questionable in view of the strong field dependence of the thermal conductance in the ordered phase and because of recent experimental investigations on phonon dynamics by inelastic x-ray scattering measurements [9]. The acoustic phonon dispersions and lifetimes do not change significantly upon cooling into the hidden-order phase, arguing against the phononic origin of the excess κ observed below T_h in URu₂Si₂. In line with this, we will show that our experimental results for CeRu₂Al₁₀, on the other hand, strongly support low-energy magnetic excitations to be at the center of these phenomena. Moreover, main features in the resistivity, thermopower, and Nernst coefficient are able to be approached by taking into account two regimes of very different charge dynamics separated by T_0 . These, again, share similarities with those of URu₂Si₂.

Polycrystalline sample of CeRu₂Al₁₀ and its nonmagnetic homologue LaRu₂Al₁₀ were prepared by arc melting the stoichiometric starting materials and following an annealing process in vacuum at 800° for one week [1]. Power xray diffraction confirms the YbFe₂Al₁₀-type (orthorhombic, space group Cmcm, No. 63) crystal structure. The obtained lattice constants are a = 9.1254 Å, b = 10.2791 Å, and c = 9.1876 Å, which are in good agreement with the reported values. Electrical resistivity $\rho(T)$, thermal conductivity $\kappa(T)$, thermopower S(T), and Nernst coefficient v(T) were measured in the physical property measurement system (PPMS, Quantum Design) between 2 K and room temperature, using a sample of typical dimension $0.5 \times 2 \times 5$ mm³. Among these, the Nernst measurements were performed on a home-design sample puck with a chip resistor of 2000 Ω as heater and a thin $(\phi = 25 \ \mu \text{m})$ chromel-AuFe_{0.07%} thermocouple for detecting the temperature gradient, as described in Ref. [10].

Figure 1(a) shows the electrical resistivity $\rho(T)$ measured in various magnetic fields applied perpendicular to electrical current, the derivative $d\rho/dT$, and the magnetoresistance MR(*T*). In agreement with previous reports [1,2], the curve of $\rho(T)$ displays a prominent maximum at $T \approx 22$ K well below T_0 . The phase transition at T_0 is, however, clearly manifested by a sharp negative extreme in both $d\rho/dT$ and MR

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FIG. 1. (a) Electrical resistivity $\rho(T)$ measured in various magnetic fields applied perpendicular to electrical current, the magnetoresistance MR_{8T} = $(\rho_{8T} - \rho_{0T})/\rho_{0T}$, and the derivative of ρ with respect to *T* for *B* = 0 T. Dotted line represents a thermal activation behavior of $\rho(T)$ observed between 30 and 80 K. (b) Hall mobility $\mu_{\rm H}$ as a function of temperature [10].

as a function of temperature. The value of MR(T) is negative above T_0 , evolving dramatically to be positive upon cooling. In other words, application of a magnetic field suppresses charge scattering events only at $T > T_0$. This points to the onset of a magnetically ordered phase at T_0 , above which spin fluctuations as scatterers of conduction electrons give rise to negative values of MR.

Given that a charge gap opens over nearly 90% of the Fermi surface slightly above T_0 [11,12], it is uncommon that $\rho(T)$ evolves from a semiconducting behavior above T_0 to be metal-like in the ordered phase. Interestingly, this unusual behavior shares some similarities to $\rho(T)$ of URu₂Si₂. Upon cooling down into the hidden-order phase, $\rho(T)$ of URu₂Si₂ passes through a maximum at a temperature below $T_{\rm h}$ and becomes more metallic at lower temperatures [13], even though the majority of the Fermi surface is gapped out. These features, observed in both compounds, apparently are related to the much larger Hall mobility $\mu_{\rm H}$ in the ordered phase relative to the paramagnetic phase [cf. Fig. 1(b)] and are consistent with the large, positive MR in the ordered phase. In a limited temperature range from above T_0 up to 80 K, $\rho(T)$ of CeRu₂Al₁₀ roughly follows a thermal activation law, with a small charge gap $E_g = 46$ K that is comparable to that estimated from NMR [11] and optical spectra [12].



FIG. 2. Thermal conductivity $\kappa(T)$ measured in different magnetic fields. The electronic contribution $\kappa_e(T)$ (crosses) was estimated from the WF law on the basis of the measured values of resistivity in zero field. The solid line represents the phononic thermal conductivity κ_{ph} of the nonmagnetic analog LaRu₂Al₁₀ and the dotted curve $\kappa(T)$ of single-crystalline CeFe₂Al₁₀ along the most heat-conductive direction (*a* axis). The dashed curve at temperatures below T_0 is an extrapolation of the measured κ for CeRu₂Al₁₀, ignoring the excess contribution below T_0 and following essentially the same *T* dependence of κ_{ph} of LaRu₂Al₁₀. By applying magnetic field, the excess heat conductance below T_0 is significantly reduced.

Figure 2 shows the measured $\kappa(T)$ for CeRu₂Al₁₀. It has two distinct maxima at T = 20 and 50 K, separated by a sharp valley right at T_0 . Such a temperature profile has been observed for both polycrystalline and single-crystalline samples [1,14] and the isoelectronic homologue CeOs₂Al₁₀ [15] with a similar phase transition. A common practice to analyze the thermal conductivity of a simple conducting solid without magnetic contribution assumes κ to be the sum of a phononic and an electronic contribution, $\kappa = \kappa_e + \kappa_{ph}$. The electronic term κ_e can be readily estimated from the Wiedemann-Franz law, $\kappa_e \rho/T = L_0$, with the Sommerfeld value of Lorenz number $L_0 \equiv \frac{\pi^2}{3} (\frac{k_B}{2})^2 = 2.44 \times 10^{-8}$ W Ω K⁻². Due to the relatively large values of resistivity of CeRu₂Al₁₀, κ_e is estimated to be negligibly small at T < 100 K, being easily ruled out as a relevant origin of the complex $\kappa(T)$ behavior.

To shed light on the origin of the double-maximum structure of $\kappa(T)$, it is instructive to first look at κ of the nonmagnetic homologue, LaRu₂Al₁₀. The phononic contribution $\kappa_{ph}(T)$ to the latter compound estimated by subtracting $\kappa_{e}(T)$, as derived from $\rho(T)$ with the aid of the WF law, from the raw data, is shown in Fig. 2. A comparison between the measured total thermal conductivity for CeRu₂Al₁₀ and $\kappa_{\rm ph}(T)$ of LaRu₂Al₁₀ reveals a good coincidence of their maxima at high temperature (40-50 K), characteristic of lattice heat conductance in a crystalline solid. Furthermore, $\kappa(T)$ of single crystalline, isoelectronic CeFe₂Al₁₀ (dotted curve, adopted from Ref. [16] for the a axis) is also shown for comparison. This compound is characterized by a stronger hybridization between conduction electrons and f states and, consequently, a nonmagnetic semiconducting ground state. Correspondingly, its $\kappa(T)$, which is dominated by the phononic



FIG. 3. The excess thermal conductivity κ_m in the ordered phase and its suppression in magnetic field, denoted as the difference of thermal conductivity measured in zero and applied magnetic fields, i.e., $\Delta \kappa = \kappa_{0T} - \kappa_B$. Note the appearance of nonzero $\Delta \kappa$ in only the ordered phase and its strong field dependence.

contribution, reveals a single, phonon-derived maximum with smaller but comparable values. These facts suggest that the maximum at T = 50 K in $\kappa(T)$ of CeRu₂Al₁₀ is most probably phononic in origin, whereas the enhancement below T_0 is due to a different mechanism. Assuming a similar temperature dependence of $\kappa_{ph}(T)$ for CeRu₂Al₁₀ to that of LaRu₂Al₁₀, one can straightforwardly extrapolate the curve of $\kappa(T)$ from above T_0 down to 2 K (dashed line in Fig. 2). The excess thermal conductivity below T_0 for CeRu₂Al₁₀, denoted as κ_m , is therefore obtained by subtracting the estimated κ_{ph} from the measured values of κ . As shown in Fig. 3(a), in the ordered phase, κ_m amounts to sizable values that are comparable to κ_{ph} .

Further evidence in support of an exotic mechanism rather than lattice vibration as origin of $\kappa_{\rm m}$ is found by applying magnetic fields: While at $T > T_0, \kappa(T)$ is field insensitive, with the onset of the ordered phase at T_0 , κ_m can be suppressed substantially by applying a magnetic field. The suppression, revealed by the difference between κ measured in zero and applied field, $\Delta \kappa = \kappa_{0T} - \kappa_{B}$, is also shown in Fig. 3. The sudden development of $\Delta \kappa$ right below T_0 and its strong field dependence point to (i) an intimate relationship of the excess thermal conductivity $\kappa_{\rm m}$ and the phase transition at T_0 and (ii) heat carriers responsible for κ_m are magnetic in origin or strongly coupled to magnetic excitations. In other words, the magnetic excitations in the ordered phase play a key role as either heat carriers or their scatterers; a pure phononic scenario for the excess κ below T_0 seems unlikely. Here one would also like to rule out structural change/distortion as a leading origin for $\kappa_{\rm m}$, given its field sensitivity.

As already mentioned, our observations on $\kappa(T)$ of CeRu₂Al₁₀ show a striking resemblance to those of URu₂Si₂: In the hidden-order phase of the latter compound, a large enhancement of κ as well as a field-induced suppression have been observed [7,8]. At $T > T_h, \kappa(T)$ is insensitive to field, similar to our observations made for CeRu₂Al₁₀. The excess κ below T_h has been interpreted as a consequence of a sudden freezing out of scattering centers mainly for heat-carrying acoustic phonons [8]. This conclusion is still under debate. Recent inelastic x-ray scattering measurements of phonon dynamics in URu₂Si₂ reveal that the acoustic phonon modes do not change significantly upon cooling into the hidden-order phase [9]. This leads to the conclusion that $\kappa_{\rm ph}$ is much less important compared to that of magnetic excitations in the low temperature phase, despite the extremely small magnetic moment [9]. By contrast, if the excess thermal conductivity in the ordered phase would mainly reflect a recovery of $\kappa_{ph}(T)$ by freezing out spin-lattice scattering events, usually one expects $\kappa_{ph}(T)$ to further increase upon applying magnetic field. For, the corresponding spin waves and fluctuations will be driven to even higher energies by field and therefore less thermally excited. This is in contrast to the observations made for CeRu₂Al₁₀ and URu₂Si₂ but is indeed the case for many multiferroic materials due to the strong spinlattice coupling [17]. Moreover, the quantitative agreement between κ_{ph} of LaRu₂Al₁₀ and the measured κ for CeRu₂Al₁₀ (Fig. 2) tends to argue against a phononic scenario for $\kappa_{\rm m}$, too.

The above inference that the magnetic excitation is the likely source of $\kappa_{\rm m}$ also finds support from neutron scattering experiments for URu₂Si₂ [18]: In spite of very tiny antiferromagnetic moments ($\sim 0.02 \ \mu_{\rm B}$) detected in the hiddenorder phase, well-defined low energy magnetic excitations throughout the Brillouin zone have been observed. Likewise, our analysis of the magnetothermal conductivity data support the same conclusion for $CeRu_2Al_{10}$, where heat transport by thermally-activated, low-energy antiferromagnetic spin waves appears to be the most likely source of $\kappa_m(T)$ below T_0 . A detailed investigation on the magnetic and phononic excitation spectra, as has been performed for URu_2Si_2 [9], may yield more conclusive information on this concern. For an insulating spin system, the spin wave scenario has been well established, and an excess heat conductance in the magnetically ordered phase has been proved to be an effective probe for low energy magnetic excitations [19]. Along the same line, the significant κ_m in CeRu₂Al₁₀ and URu₂Si₂ is believed to benefit from the low carrier density of the ordered phase in these compounds. However, it remains to be unraveled why a magnetic contribution to $\kappa(T)$ in a system with very small magnetic moments can be substantially large. In view of the high similarity of the thermal conductivity and close magnetic ordering temperature of CeRu₂Al₁₀ and CeOs₂Al₁₀ [15], it seems reasonable to believe that the above discussion applies to the latter compound as well.

The aforementioned discussion is based on the argument that in the current systems magnetic spin waves act mainly as heat carriers rather than scatterers. This is reasonable considering the abrupt increase of thermal conductivity upon onset of the low-temperature phase. We note however that, as far as field suppression of κ is concerned, a phononic thermal conductance cannot be fully excluded from the excess thermal conductivity κ_m . Under certain circumstances, e.g., when the system is in the vicinity of a field-induced magnetic instability [20] or experiences a resonant scattering of phononic and magnetic excitations [21], a suppression of κ_{ph} occurs in a corresponding field region. Further experimental and theoretical work is needed to verify whether this can partially account for the field dependence of $\Delta \kappa$ below T_0 .



FIG. 4. (a) Thermopower S(T) measured in different magnetic fields for CeRu₂Al₁₀. The curve of $\rho(T)$ is also shown in order to highlight the similarity of the sharp peak at 22 K in both S(T) and $\rho(T)$. Inset: The magnetothermopower defined $\Delta S = S_{\text{8T}} - S_{\text{0T}}$. (b) Nernst coefficient ν measured in different fields shown as $-\nu \text{ vs } T$.

In Figs. 4(a) and 4(b) we show the thermopower S(T) and Nernst coefficient v(T) measured in different magnetic fields for CeRu₂Al₁₀. S(T) measured at B = 0 T is characterized by two broad maxima at T = 7 K and 180 K, together with a sharp peak emerging in between, at $T \approx 22$ K, in agreement with previous reports [1,6]. On the other hand, $|\nu(T)|$ is dramatically enhanced by two orders of magnitude with the onset of the ordered phase below T_0 . Except for the sharp peak at 22 K, a double-maximum feature of S(T) had been frequently observed for Kondo systems, e.g., CeRu₂Si₂ [22], reflecting the Kondo scattering on the crystalline-electric-field (CEF) derived Kramers ground-state and excited doublets. Indeed, the first excited doublet of CeRu₂Al₁₀ in the orthorhombic CEF is about 30 meV above the ground state [23], in rough accordance with the position of the high temperature maximum of S(T). Interestingly, the values of S(T) remain the same magnitude while the carrier concentration changes by two orders of magnitude for the two phases separated by T_0 [1]. This is the case for URu_2Si_2 [24], too. For Kondo systems, such an observation is not surprising because S is strongly dependent on the Kondo scattering rather than the electronic density of states (DOSs) of the conduction band [25]. Upon applying magnetic fields, at T < 20 K the values of S(T) commence to become reduced, whereas S(T) remains unchanged at higher temperatures, as revealed by $\Delta S(=S_{0T} - S_{8T})$ as a function of temperature [Fig. 4(a), inset]. These distinctly different field responses of thermopower point to two different Kondo energy scales below and above T_0 , related to the abrupt change of the quasiparticle DOSs by opening of a charge gap. S(T) varies with temperature sublinearly below the low-temperature maximum, with an initial slope $S/T = 4.8 \ \mu V/K^2$ at B = 0 T. Given the electronic specific-heat coefficient $\gamma = 24.5 \ mJ/mol \ K^2$ in the ordered phase [2], the dimensionless ratio of S/T and γ , i.e., $q = S/\gamma T$, amounts to 18.8 for CeRu₂Al₁₀. This value, as well as q = 4.5 for URu₂Si₂ [26], is larger than $q \ (\approx 1)$ of most heavy-fermion metals, consistent with the low-carrier nature of their ordered phases.

The sharp S(T) peak at $T \approx 22$ K appears to signify the temperature regime where both $\mu_H(T)$ and $\nu(T)$ [Fig. 4(b)] show pronounced changes. Due to its field insensitivity this peak cannot be ascribed to a magnon-drag effect associated with electron-spin wave interaction. We recently have demonstrated for the electron-doped skutterudite CoSb₃ that an anomalous thermopower can stem from a dramatic change of charge mobility with respect to temperature [10]. While the thermopower is commonly referred to as a measure of energy dependence of the electronic DOSs at the Fermi level, it may well derive from a significantly energy-dependent relaxation time τ as evidenced by a largely temperature-dependent $\mu_{\rm H}$. Following this description, the S(T) peak at 22 K can be qualitatively described by the derivative of Hall mobility, $d\mu_H/dT$. For details, see the supplementary information of Ref. [10]. The fact that this anomalous S(T) peak sits right on top of the $\rho(T)$ maximum [cf. Fig. 4(a)] lends further support to this argument. Noticeably, the absolute value |S(T)|of URu₂Si₂ increases abruptly below T_h as well [24], which may be partially derived from the strong variation of $\mu_{\rm H}(T)$ as discussed above. When magnetic field is applied, even a two-maximum structure in S(T) is observed below $T_{\rm h}$ [24], mimic to the case of CeRu₂Al₁₀.

The Nernst coefficient $|\nu(T)|$, which increases by more than two orders of magnitude with the onset of the lowtemperature phase in both the current system [Fig. 4(b)] and URu₂Si₂ [24,27], deserves special attention as well. Within the Boltzmann theory, one indeed expects a large Nernst signal from high mobility charge carriers: In a first-order approximation, $\nu \propto \frac{k_{\rm B}T}{\epsilon_{\rm F}}\mu_{\rm H}$, with $k_{\rm B}$ being Boltzmann constant and $\epsilon_{\rm F}$ the Fermi energy. Nevertheless, the microscopic mechanism for large Nernst signal in these compounds may not be that straightforward. For URu₂Si₂, additionally, the large Nernst response has been discussed as a fingerprint of possible chiral or Berry-phase fluctuations associated with the broken time-reversal symmetry of the superconducting order parameter [27]. Finally, it is instructive to mention that, apart from transport signatures, analogies between the two systems have also been found by other probes, such as optical conductivity, which exhibits a gap structure with a mysterious charge excitation peak [12], and NQR/NMR spectra, which reveal a similar nuclear-spin lattice relaxation rate for both compounds [11].

To summarize, we have investigated various magnetotransport properties of the Kondo semiconducting compound CeRu₂Al₁₀ and stressed their analogies with those of the hidden-order compound URu₂Si₂. With the onset of the ordered phase at $T_0 = 27$ K, CeRu₂Al₁₀ experiences a dramatic change of charge dynamics exhibiting very different charge mobilities below and above T_0 . This has profound effects on the electrical and thermoelectrical responses, leading to an additional peak in both $\rho(T)$ and S(T), as well as an enhanced Nernst coefficient $\nu(T)$, resembling the case of URu₂Si₂. Most significantly, in the ordered phase, thermal conductivity exhibits an excess contribution presumably derived from lowenergy magnetic excitations and subject to field suppression, in strong parallel to URu₂Si₂ as well. Given the Kondo semiconducting behavior above T_0 , which usually involves a

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nonmagnetic ground state, it is surprising that this compound shows magnetic order with small moments at a surprisingly high temperature. Like the hidden order phase in URu₂Si₂, where the tiny antiferromagnetic moments cannot be the main order parameter below T_h , the low-temperature magnetic phase in CeRu₂Al₁₀ remains elusive and badly calls for further work.

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